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### Abstract

Recently, it has been shown that several artists' pigments will fade in the absence of light if exposed to atmospheric ozone at the levels found in Los Angeles photochemical smog. In the present study, a large number of organic artists' watercolors have been examined to further assess the scope of this fading hazard to works of art. Twenty-seven organic artists' watercolors were exposed to  $0.31 \pm 0.12$  ppm  $O_3$  at  $46 \pm 6\%$  RH for 90 days. It was found that all of the watercolors containing alizarin-derived pigments (1,2 dihydroxyanthraquinone lakes) are susceptible to fading in the presence of ozone. A number of pigment blends containing at least one ozone-fugitive chemical component were found to change their apparent color after exposure to ozone. Several organic red pigments were identified that are much more ozone-resistant than the alizarin lakes, including quinacridone reds and BON arylamide reds. This information may aid artists in selecting more durable substitutes for the ozone-fugitive alizarin lakes.

## 1. INTRODUCTION

Extended exposure to air pollution has long been recognized as a possible cause of damage to works of art and cultural properties. Early studies linked much of this damage to sulfur-containing pollutants emitted from combustion sources (1). To this day, the caustic effects of acidic air pollutants on leather and paper (2-4), stone (5-7), bronze (8), stained glass (9), lead white pigments (10), and other materials are being documented.

Awareness of the outdoor air quality in modern metropolitan areas has led to growing concern over the hazards from another source: photochemical smog. The composition and chemistry of this enormously complicated mixture of gases, aerosols, and particulate matter slowly are being unraveled after extensive research (11). One of the major components of photochemical smog, ozone ( $O_3$ ), has been clearly established as a powerful oxidizing species, capable of causing deterioration of many materials, including textile fibers (12) and dyes (13), rubber (14), and the binders found in exterior paints (15). The risk of ozone damage is not confined solely to urban areas, for the long-term average ozone concentrations in cities like Los Angeles are only slightly higher than in many remote areas (16,17).

The issue of the intrusion of outdoor ozone into museums was first raised by Thomson (18,19). Very recently, measurements made at two different museums confirmed the presence of indoor ozone at concentrations 60% to 70% as high as outdoor levels (20,21). In contrast, two other museums that possess activated carbon filters on the inlets to their air conditioning systems registered very low indoor ozone levels (20).

Having established that ozone can reach significant levels inside a museum building, proper assessment of the risk to museum collections requires a knowledge of the ozone sensitivity of the materials commonly used by artists and conservators. Much of the documentation of ozone attack has been accumulated by the rubber and textile industries, and the ozone resistance of some museum materials will be found in that trade literature. Those same studies, however, indicate that the ozone reactivity of a particular chemical

compound (a textile dye, for example) is dependent on the entire system (the fiber-mordant-dye combination) and its physical state (22), a phenomenon that also has been observed in lightfastness tests (23). This suggests that screening tests should be performed on the specific materials found in museum collections in order to determine their individual sensitivities to atmospheric pollutants. In addition to targeting fugitive materials requiring appropriate precaution, this procedure also would identify materials that are particularly ozone-resistant and, it is hoped, serve as a guide towards the production of more permanent works.

As a first step toward this end, a pilot experiment was conducted in our laboratory in 1982 (20). In that exploratory test, hereafter referred to as Experiment I, samples of eleven organic Winsor and Newton artists' watercolors on paper and two Japanese woodblock prints were exposed to an atmosphere containing 0.40 parts per million (ppm) ozone at 50% relative humidity in the absence of light for 95 days. Three of the watercolors, two containing an alizarin (1,2 dihydroxyanthraquinone) lake pigment and the other a blend containing a triphenylmethane (Basic Violet 14) lake, faded dramatically. A fourth watercolor, also containing an alizarin-based pigment, faded to a lesser extent. Also showing definite fading were the yellow colorants used in the Japanese woodblock prints. Microchemical tests performed on one of the woodblock prints suggested that the yellow colorant was either gamboge or turmeric. Complete details of the results of Experiment I are reported elsewhere (20).

In this paper, a second ozone study is described in which the experimental protocol of Experiment I was used to survey the fading characteristics of a larger number of Winsor and Newton tube watercolors. Two considerations motivated the selection of these samples. First, a wide variety of organic pigment classes was included so that their ozone reactivities could be compared to those of the sensitive pigments identified in Experiment I (alizarin lake, triphenylmethane lake, and either gamboge or turmeric). Secondly, a series of watercolors was chosen, all of which contain either alizarin lake or triphenylmethane lake and represent various pigment blends and/or lake formulations.

By comparing the fading behavior of these preparations, we hoped to illustrate the range of sensitivity to be found within one manufacturer's alizarin- and triphenylmethane-derived pigments.

## 2. EXPERIMENTAL PROGRAM

The complete list of twenty-seven Winsor and Newton watercolors is given in Table I. Samples were prepared by brushing a dilute aqueous suspension of each watercolor chosen onto 5 cm by 10 cm strips of watercolor paper (Bockingford 140 lb cold-pressed, pH neutral). Fifty-one samples were created, giving two examples (one lightly tinted and one more saturated) for twenty-four of the twenty-seven watercolors. These paper strips then were cut in half to form two identical swatches. One set was kept in sealed opaque envelopes for the duration of the experiment to serve as unexposed controls. The second set of paper samples was mounted, twelve to a page, onto backing sheets of watercolor paper using picture corners, and these sheets were hung adjacent to the inside walls of the exposure chamber with the color patches facing the open interior of the box.

A schematic diagram of the ozone exposure apparatus is shown in Figure 1. Air was pumped at a rate of 2.2 liters/minute through a purification system designed to remove preexisting pollutants. The first stage of this filtration system consisted of a packed bed of fresh Purafil (alumina impregnated with potassium permanganate) to remove  $\text{SO}_2$  and  $\text{NO}_2$ . A canister of activated carbon was employed to remove ozone and reactive hydrocarbons from the air stream. A particulate filter then removed any aerosol present and also captured any fragments from the Purafil and activated carbon beds that might have become entrained in the flow. The filtered air was next re-ozonated by an ultraviolet light source (Ultra Violet Products, Inc., Pen-ray Lamp) and finally dispersed into the exposure chamber through a perforated Teflon tube. The air within the exposure chamber was stirred continuously, and the ozone concentration within the chamber was measured and recorded continuously using an REM chemiluminescent ozone monitor and strip chart recorder.

The watercolor samples were exposed to  $0.31 \pm 0.12$  ppm ozone in the absence of light for 90 days at a temperature of  $26 \pm 3^\circ\text{C}$  and a relative humidity of  $46 \pm 6\%$ . Color differences between the ozone-exposed samples and the unexposed controls were determined by visual color matching under Sylvania Warmlight fluorescent lighting. The Munsell System of color notation was used to define each sample by hue, value, and chroma (24). In addition, diffuse reflectance spectra between 400 and 700 nm were measured using a Varian Cary model 219 spectrophotometer equipped with an integrating sphere reflectance attachment.

### 3. RESULTS

The Munsell color matches for the control and the ozone-exposed watercolor samples are listed in Table I. Also shown are the chemical compositions of the tube watercolors tested as reported in the Winsor and Newton product literature (25). The watercolors have been categorized according to hue, and the results of the exposure tests for each category are described.

#### 3.1. Red Pigments

Previous work by Shaver et al. during Experiment I showed that the red alizarin lake pigments alizarin crimson and crimson lake fade rapidly when exposed to atmospheric ozone (20). In order to establish correspondence between Experiment I and the present work, the ozone-fastness of these two pigments was tested. The previous findings were reconfirmed, both the alizarin crimson and crimson lake samples fading as before. As shown in Figure 2, the ozone-exposed crimson lake sample exhibits increased reflectance relative to the control sample, resulting in the faded appearance illustrated in Figure 3.

During Experiment I, a third alizarin lake watercolor, purple lake, showed a barely detectable color change, and some uncertainty remained about whether or not that pigment would fade in a manner similar to alizarin crimson and crimson lake. As shown in Figure 4, that doubt now has been erased. The fading of purple lake is still less extreme

than that of alizarin crimson and crimson lake, but it is clearly measurable during this experiment, both by the spectrophotometer and by a human observer.

Two other watercolors reported to contain alizarin lakes are alizarin carmine and brown madder. As shown in Figure 5, alizarin carmine fades upon reaction with ozone, but again the extent of fading is less severe than that observed for alizarin crimson and crimson lake. Brown madder has an initial spectral reflectance that is quite different from the red alizarin lakes, yet it too fades when exposed to ozone, as is seen in Figure 6.

The Winsor and Newton watercolor carmine contains a red pigment prepared as a lake from natural cochineal, which is a highly substituted anthraquinone ring structure as seen in Figure 7. Its appearance is very similar to the alizarin lakes, as can be seen by comparing the spectral reflectance curve of Figure 8 to that of alizarin carmine in Figure 5. During Experiment I, the carmine sample tested was resistant to ozone attack to the extent that no definite change in color could be demonstrated. In the present study, two samples of carmine were prepared and tested. The Munsell color matches shown in Table I indicate a very slight fading of both carmine samples. However, the spectrophotometer results show the slight fading of one sample (see Figure 8) but no measurable fading of the second sample.

A quinacridone, Pigment Violet 19 (CI 46500), is the red colorant in three of the watercolors tested: permanent rose, permanent magenta, and rose doré. The structure of quinacridone is given in Figure 7. All of these samples remained virtually unchanged after ozone exposure, as illustrated in Figure 9 with the spectral reflectance curve of one of the permanent rose samples.

Also relatively ozone-resistant were the watercolors rose carthame and scarlet lake, which both contain a BON arylamide red, and bright red, a blend of chlorinated parared (CI 12085) plus arylamide yellow (CI 11680). Samples of these watercolors showed almost no change in Munsell match upon ozone exposure. The reflectance spectra of control and exposed samples indicate no change within instrumental resolution for the rose carthame and less than

2% increase in reflectance in the blue and green for scarlet lake.

### 3.2. Yellow Pigments

The known gamboge watercolor sample exposed to ozone during Experiment I showed no definite color change. In the present study, a sample of Winsor and Newton's genuine gamboge tube watercolor (Natural Yellow 24) was prepared and exposed to ozone. This gamboge sample showed a very slight increase in reflectance, a difference too small to be certain that it was due to the ozone exposure rather than to a slight non-uniformity between the exposed and control sample strips.

Synthetic yellow pigments examined during the present experiment include Winsor and Newton's new gamboge, "Indian yellow," and Winsor yellow. Each is based on arylamide yellow (CI 11680). All show small increases in reflectance (on the order of a few percent) after ozone exposure, like that seen in Figure 10. A change of this magnitude is difficult to detect visually: the Munsell matches given in Table I show little or no change in color as a result of 90 days' exposure to 0.31 ppm ozone.

### 3.3. Green Pigments

Winsor green, derived from chlorinated copper phthalocyanine green (CI 74260), is one of the most ozone-resistant organic pigments tested during this experiment. As seen in Figure 11, there are no detectable differences between the spectral reflectance curves of the control and the exposed Winsor green samples. Olive green light, formulated from chlorinated copper phthalocyanine and raw sienna (CI 77492), also shows no detectible color change after ozone exposure. The spectrophotometer reflectance curves for Hooker's green light and Hooker's green dark, both blends of chlorinated copper phthalocyanine and a ferrous beta naphthol derivative (similar to CI 11020), show signs of slight fading in three out of four samples tested. The extent of fading was small, as is seen in the Munsell data of Table I. The watercolor "sap green" contains the same colorants as Hooker's green



with the addition of arylamide yellow. The reflectance spectra show that "sap green" faded slightly in a manner similar to the Hooker's green samples, and again the visual extent of fading was small when specified by Munsell color matching.

Prussian green in the Winsor and Newton watercolor line contains a blend of arylamide yellow and Prussian blue. Ozone-exposed Prussian green samples show practically the same spectral reflectance as the unexposed controls. In the case of one sample, the Munsell color match suggests a very small change, while the Munsell match of the second sample shows no difference between exposed and control samples.

#### 3.4. Blue, Gray, and Purple Pigments

Several watercolors called by traditional names now are formulated as a blend of pigments. Examples from the Winsor and Newton line are the tube watercolors "indigo," "Payne's gray," and "mauve." Winsor and Newton's "indigo" watercolor pigment is no longer made from natural indigo, but rather is formulated from lamp black, copper phthalocyanine, and an alizarin lake (25). As seen in Figure 12, the ozone-exposed "indigo" sample faded slightly in this experiment. Lamp black is almost certainly stable in the presence of ozone, and the alizarin lake is expected to be ozone-sensitive. The reactivity of the copper phthalocyanine has not yet been determined. We note that much of the increased reflectance of the faded "indigo" occurs in the 500 nm range, suggesting the reaction of the pigment which absorbs in that spectral region. That pigment would be the alizarin lake, whose extreme sensitivity was observed for the red watercolors.

According to Brommelle's summary of the Russell and Abney report on the lightfastness of artists' watercolors (26), in the 1880's Winsor and Newton made Payne's gray by blending natural indigo, crimson lake, and carbon black. As in the case of their "indigo" watercolor, since that time Winsor and Newton have reformulated the material that they sell as "Payne's gray." The sample tested here is reportedly a mixture of alizarin lake, lamp black, Prussian blue, and ultramarine (25). As shown in Figure 13, "Payne's gray" faded significantly during this experiment, with increased

reflectance in the blue and green, giving a lighter and bluer shade. Again, the increased reflectance in the 500 nm region suggests that the fading is at least in part a result of the reaction of the alizarin lake component of the blend. The reflectance at the far red end of the spectrum (700 nm) is left unchanged after ozone exposure, suggesting that the inorganic blue pigments contained in the blend (Prussian blue and ultramarine) probably have not been affected.

Winsor and Newton's "mauve" watercolor is unrelated to the traditional aniline dye, but is instead reportedly a blend of a triphenylmethane lake (a lake of Basic Violet 14) plus copper phthalocyanine (25). It was observed in Experiment I that ozone exposure causes a color change from purple to a lighter blue shade. This result was duplicated in the present study. Figure 14 shows the increase of spectral reflectance in the blue region caused by ozone exposure, and the appearance of the color shift is illustrated in Figure 15. The observed color change suggests that the red triphenylmethane lake is reacting with ozone, leaving the pigment blend slightly richer in the blue component (copper phthalocyanine, in this case).

In order to explore this possibility further, Winsor violet, another pigment blend including a triphenylmethane lake, was tested. The result of the ozone exposure suggests that the Winsor violet sample may have faded slightly, but there was little indication of a color shift as pronounced as that observed for the "mauve" sample.

#### 4. DISCUSSION

The ozone sensitivity of alizarin lake pigments has been demonstrated to be a general property within the Winsor and Newton watercolor line. The five alizarin-derived red pigments tested and the two pigment blends, "indigo" and "Payne's gray," reported to contain alizarin lake, all proved fugitive upon exposure to ozone. The detailed chemical compositions of these pigments are not clearly stated by the manufacturer, so it is not known whether the different shades are produced by altering the lake formulation or by the addition of other colorants. For the present, however, it would be prudent to consider the entire

family of alizarin-based pigments to be ozone-fugitive.

The observed reactivity of alizarin lake and the rapid ozone fading of some anthraquinone textile dyes (27) seem to be unreliable indicators of the ozone sensitivity of other anthraquinone pigments. For example, in Experiment I and in this study the samples of carmine, a lake of a highly substituted anthraquinone ring system, proved to be much less ozone-fugitive than the alizarin lake pigments (see Figure 7). Presumably the substituents on the anthraquinone molecule or the physical structure of the lake can alter the rate of ozone attack to such an extent that over the duration of an ambient ozone exposure equivalent to that represented by these experiments (roughly six years inside an unprotected museum in Los Angeles) (20), carmine faces a smaller risk than alizarin crimson.

Similarly, the test results of the "mauve" and Winsor violet watercolors illustrate the difficulty of predicting the ozone sensitivity of a pigment blend. In addition to the ozone reactivity of its individual components, many other factors apparently can contribute to the color change upon ozone exposure: the relative amounts of the pigments in the blend, the relative colorant strengths of the components, and interactions between the pigments or between pigments and ozone degradation products all could be important. At this time there is very little information available about procedures for producing the "mauve" blend and the Winsor violet watercolors, so their very different ozone sensitivities remain unexplained.

Another unanswered question is the identity of the ozone-fugitive organic yellow colorant from the Japanese woodblock print of Experiment I. During that study, small amounts of a strong base (NaOH) were applied to a yellow-pigmented area of one of the prints, producing a color change from yellow to orange that is characteristic of either gamboge or turmeric (28,29). The gamboge tested in that experiment and in the present work, however, proved to be relatively ozone-resistant. The ozone-fastness of curcumin, the colorant in turmeric, is being examined during a third experiment currently underway.

## 5. CONCLUSION

Previous work in lightfastness testing of dyes and pigments and in pollution testing of textile dyes leads one to expect that the ozone-fastness of artists' pigments will be dependent on many factors, including chemical composition, manufacturing process, and the physical system in which the colorant is used (e.g., binders). In this, the second in a series of tests, we have focused on the organic pigments from the Winsor and Newton line of tube watercolors. Among the pigments which faded drastically during this experiment (and thus face potential damage from a few years of exposure in a museum environment) are all of the red pigments which contain an alizarin lake. The pigment blends "indigo," "Payne's gray," and "mauve" also displayed sensitivity toward ozone. Several organic pigments were identified that resist ozone exposure much better than the alizarin lakes, and these include quinacridone and BON arylamide reds, arylamide yellows, and chlorinated copper phthalocyanine green.

Control of indoor ozone levels provides one approach to the reduction of damage due to photochemical smog. It has been shown that the indoor ozone concentrations in museums that lack a chemically protected air conditioning system can be as high as 50% to 70% of the ozone concentration in the outdoor air that supplies the building ventilation system. Activated carbon filters on the inlets to building air conditioning systems have been proven to be effective in controlling indoor ozone levels. Because long-term average ozone levels as high as those in Los Angeles prevail throughout the northern hemisphere, we believe that precautions should be considered in order to protect museum collections from damage due to atmospheric ozone.

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Table I. Chemical Composition of Organic Artists' Pigments and Color Changes Noted After 90 Days of Exposure to 0.31 O<sub>3</sub>.

Sample Identification	Pigment Name	Chemical Composition	Munsell Notation	
Red Pigments			Color of Control Sample hue value/chroma	Color of Ozone Exposed Sample hue value/chroma
Alizarin Lakes				
002 E2P3	Alizarin Crimson	1,2 dihydroxyanthraquinone lake	9RP 6.6/10.8	7.5RP 8.6/5.2
002 E2P5	Alizarin Crimson	1,2 dihydroxyanthraquinone lake	8.5RP 6.3/11.6	7.5RP 8.4/5.6
017 E2P2	Crimson Lake	1,2 dihydroxyanthraquinone lake	7RP 6.0/12.0	5RP 7.2/8.0
017 E2P5	Crimson Lake	1,2 dihydroxyanthraquinone lake	8RP 5.5/12.8	5.5RP 7.4/8.8
038 E2P3	Purple Lake	1,2 dihydroxyanthraquinone lake	7.5RP 6.2/8.0	7.5RP 6.8/6.4
038 E2P5	Purple Lake	1,2 dihydroxyanthraquinone lake	7.5RP 5.7/8.0	7RP 6.5/6.8
001 E2P2	Alizarin Carmine	1,2 dihydroxyanthraquinone lake	10RP 5.8/10.0	10RP 6.2/10.0
001 E2P5	Alizarin Carmine	1,2 dihydroxyanthraquinone lake	10RP 5.7/10.8	10RP 6.5/9.2
007 E2P2	Brown Madder	1,2 dihydroxyanthraquinone lake	6.25R 6.6/9.2	9R 7.3/7.6
007 E2P5	Brown Madder	1,2 dihydroxyanthraquinone lake	7R 5.8/10.4	8.5R 6.7/8.4
Natural Reds				
094 E2P3	Carmine	Cochineal	9.5RP 6.5/5.6	9.5RP 6.7/9.2
094 E2P5	Carmine	Cochineal	8.75RP 5.8/12.8	8.75RP 5.9/12.4
Quinacridone Reds				
073 E2P2	Permanent Magenta	Violet 19 - a quinacridone	2.5RP 5.0/10.8	2.5RP 5.0/10.8
073 E2P4	Permanent Magenta	Violet 19 - a quinacridone	1.5RP 5.4/10.0	1.25RP 5.5/10.0
075 E2P2	Permanent Rose	Violet 19 - a quinacridone	5RP 7.3/10.8	5RP 7.3/10.8
075 E2P5	Permanent Rose	Violet 19 - a quinacridone	5RP 7.4/10.8	5RP 7.4/10.8
089 E2P2	Rose Doré	Violet 19 plus Yellow 3	2.5R 7.8/6.8	2.5R 7.8/6.8
089 E2P5	Rose Doré	Violet 19 plus Yellow 3	2.5R 8.0/6.4	2.5R 8.0/6.4
BON-Arylamides				
044 E2P2	Scarlet Lake	Red 10 - a BON arylamide	4.5R 7.4/10.4	4.5R 7.4/10.4
044 E2P5	Scarlet Lake	Red 10 - a BON arylamide	4.5R 7.0/10.4	4.5R 7.0/10.4
076 E2P2	Rose Carthame	BON arylamide & eosine lake	5.5R 6.7/10.8	5.5R 6.7/10.8
076 E2P5	Rose Carthame	BON arylamide & eosine lake	4R 7.0/10.0	4R 7.0/10.0
Other Reds				
006 E2P2	Bright Red	Chlorinated para red and arylamide yellow	10.5R 7.0/10.8	10.5R 7.1/10.8
006 E2P5	Bright Red	Chlorinated para red and arylamide yellow	10R 6.5/13.2	10R 6.6/13.2
Yellow Pigments				
Natural Yellows				
069 E2P1	Gamboge	Gambogic acid	6Y 9.0/5.6	6.5Y 9.0/5.6
Arylamide Yellows				
020 E2P1	New Gamboge	Arylamide yellow & toluidine red	7Y 9.3/4.8	7Y 9.3/4.8
020 E2P4	New Gamboge	Arylamide yellow & toluidine red	3Y 8.7/12.4	3Y 8.7/12.4
024 E2P1	"Indian Yellow"	Arylamide yellow	5Y 9.2/5.6	5Y 9.5/5.6
058 E2P1	Winsor Yellow	Arylamide yellow	7.5Y 9.2/6.4	7.5Y 9.2/6.4
Green Pigments				
Containing chlorinated copper phthalocyanine				
055 E2P1	Winsor Green	Chlorinated copper phthalocyanine	9.5G 7.7/6.8	9.5G 7.8/6.8
055 E2P4	Winsor Green	Chlorinated copper phthalocyanine	10.5G 6.2/10.8	10.5G 6.2/10.8
021 E2P1	Hookers Green - Dark	Chlorinated copper phthalocyanine & a ferrous beta naphthol derivative	3.5G 6.8/8.0	3.5G 7.0/8.0
021 E2P4	Hookers Green - Dark	Chlorinated copper phthalocyanine & a ferrous beta naphthol derivative	5G 6.5/7.2	5G 6.7/6.8
022 E2P1	Hookers Green - Light	Chlorinated copper phthalocyanine & a ferrous beta naphthol derivative	10.5GY 7.5/7.2	10GY 7.6/6.8
022 E2P4	Hookers Green - Light	Chlorinated copper phthalocyanine & a ferrous beta naphthol derivative	2G 6.2/7.6	2G 6.5/7.6
033 E2P1	Olive Green	Chlorinated copper phthalocyanine & raw Sienna	1.5GY 8.2/5.6	1.5GY 8.4/5.6
033 E2P4	Olive Green	Chlorinated copper phthalocyanine & raw Sienna	2.5GY 7.8/6.0	2.5GY 7.8/6.0
043 E2P1	"Sap Green"	Chlorinated copper phthalocyanine, a monoazo acid yellow lake, & a ferrous beta naphthol derivative	7.5GY 8.4/6.8	7.5GY 8.6/6.4
043 E2P4	"Sap Green"	Chlorinated copper phthalocyanine, a monoazo acid yellow lake, & a ferrous beta naphthol derivative	8GY 7.4/8.4	8GY 7.6/8.4
Other Greens				
037 E2P1	Prussian Green	Arylamide Yellow and Prussian Blue	1B 5.3/4.8	1B 5.3/4.8
037 E2P4	Prussian Green	Arylamide Yellow and Prussian Blue	10BG 5.9/4.8	10BG 6.0/4.4
Blue and Purple Pigments				
Blends containing an Alizarin Lake				
025 E2P1	"Indigo"	1,2 dihydroxyanthraquinone lake; lamp black and copper phthalocyanine	10B 4.7/4.8	7.5B 4.7/4.4
025 E2P4	"Indigo"	1,2 dihydroxyanthraquinone lake; lamp black and copper phthalocyanine	10B 4.7/5.6	7.5B 5.4/5.2
034 E2P1	"Payne's Gray"	1,2 dihydroxyanthraquinone lake; Prussian Blue; lamp black and ultramarine	10B 6.4/3.2	7.5B 6.5/3.6
034 E2P4	"Payne's Gray"	1,2 dihydroxyanthraquinone lake; Prussian Blue; lamp black and ultramarine	10B 6.2/3.6	7.5B 6.4/3.6
Blends containing Triphenylmethane Lake				
030 E2P2	"Mauve"	Triphenylmethane Lake (lake of Basic Violet 14), and copper phthalocyanine	3P 6.5/8.8	5PB 7.4/6.0
030 E2P4	"Mauve"	Triphenylmethane Lake (lake of Basic Violet 14), and copper phthalocyanine	2.5P 5.5/10.0	5PB 7.2/6.4
057 E2P2	Winsor Violet	Triphenylmethane Lake (lake of Basic Violet 14), plus a lake of Acid Blue 45.	5P 6.5/8.4	5P 6.7/8.4
057 E2P4	Winsor Violet	Triphenylmethane Lake (lake of Basic Violet 14), plus a lake of Acid Blue 45.	5P 5.0/10.8	5P 5.7/10.4

## FIGURE CAPTIONS

- Figure 1            Experimental Apparatus
- Figure 2            Reflectance spectrum of ozone-exposed crimson lake vs. reflectance spectrum of an unexposed control sample
- Figure 3            Appearance of ozone-exposed crimson lake vs. the appearance of an unexposed control sample
- Figure 4            Reflectance spectrum of ozone-exposed purple lake vs. reflectance spectrum of an unexposed control sample
- Figure 5            Reflectance spectrum of ozone-exposed alizarin carmine vs. reflectance spectrum of an unexposed control sample
- Figure 6            Reflectance spectrum of ozone-exposed brown madder vs. reflectance spectrum of an unexposed control sample
- Figure 7            The chemical structure of several coloring agents.
- Figure 8            Reflectance spectrum of ozone-exposed carmine vs. reflectance spectrum of an unexposed control sample
- Figure 9            Reflectance spectrum of ozone-exposed permanent rose vs. reflectance spectrum of an unexposed control sample
- Figure 10           Reflectance spectrum of ozone-exposed Winsor yellow vs. reflectance spectrum of an unexposed control sample
- Figure 11           Reflectance spectrum of ozone-exposed Winsor green vs. reflectance spectrum of an unexposed control sample

- Figure 12      Reflectance spectrum of ozone-exposed Winsor and Newton's synthetic "indigo" vs. reflectance spectrum of an unexposed control sample
- Figure 13      Reflectance spectrum of ozone-exposed Payne's gray vs. reflectance spectrum of an unexposed control sample
- Figure 14      Reflectance spectrum of ozone-exposed Winsor and Newton's "mauve" vs. reflectance spectrum of an unexposed control sample
- Figure 15      Appearance of ozone-exposed Winsor and Newton's "mauve" vs. appearance of an unexposed control sample

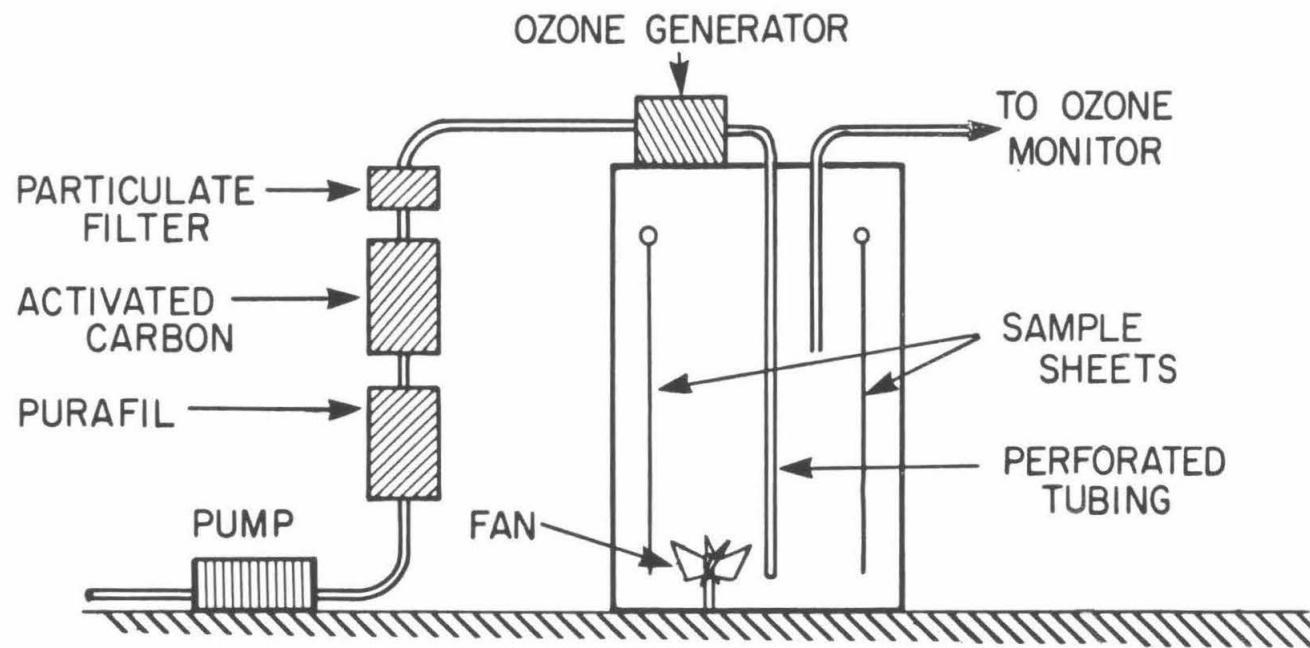


FIGURE 1

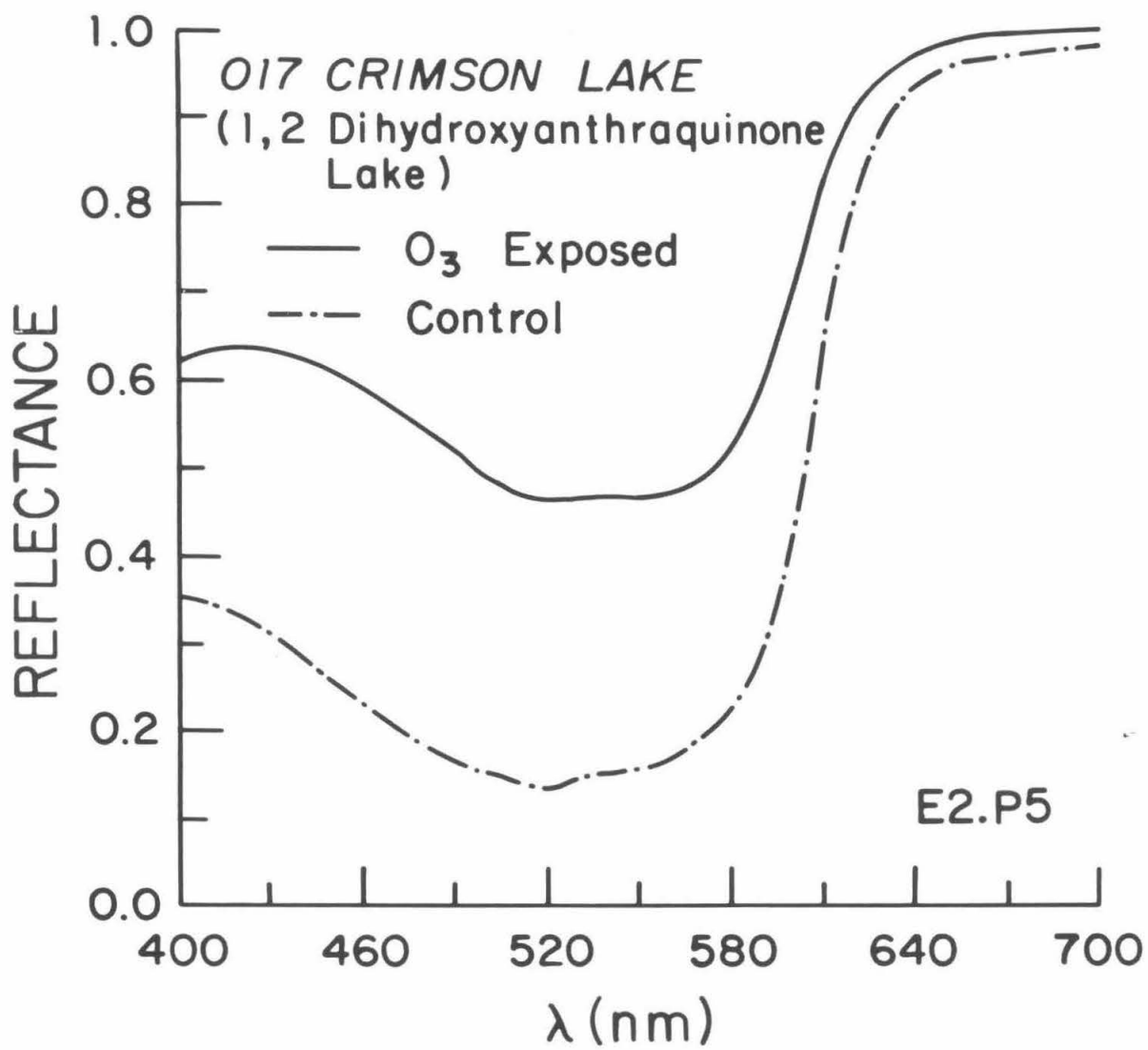


FIGURE 2

FIGURE 3 is a color slide (enclosed)

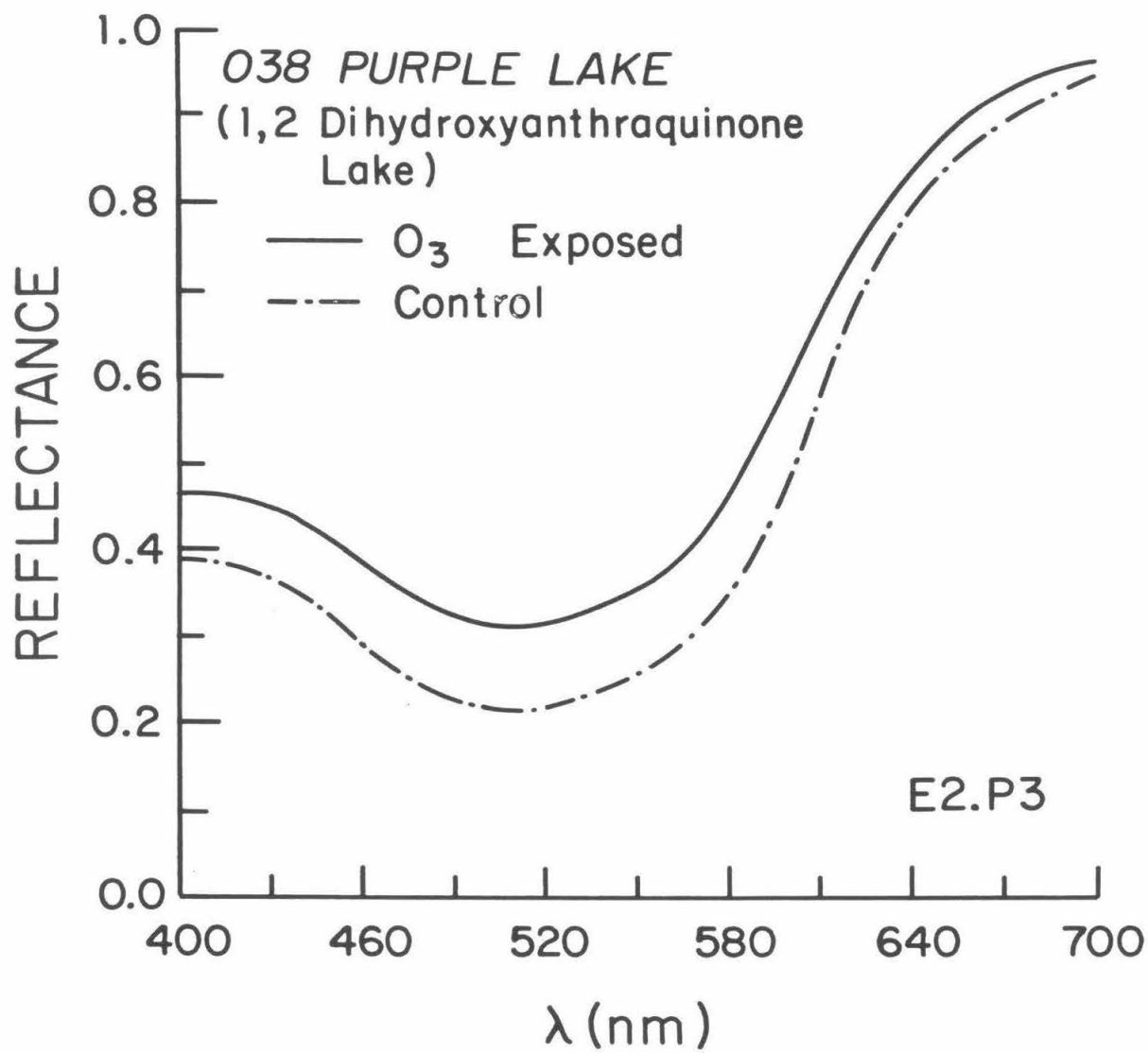


FIGURE 4

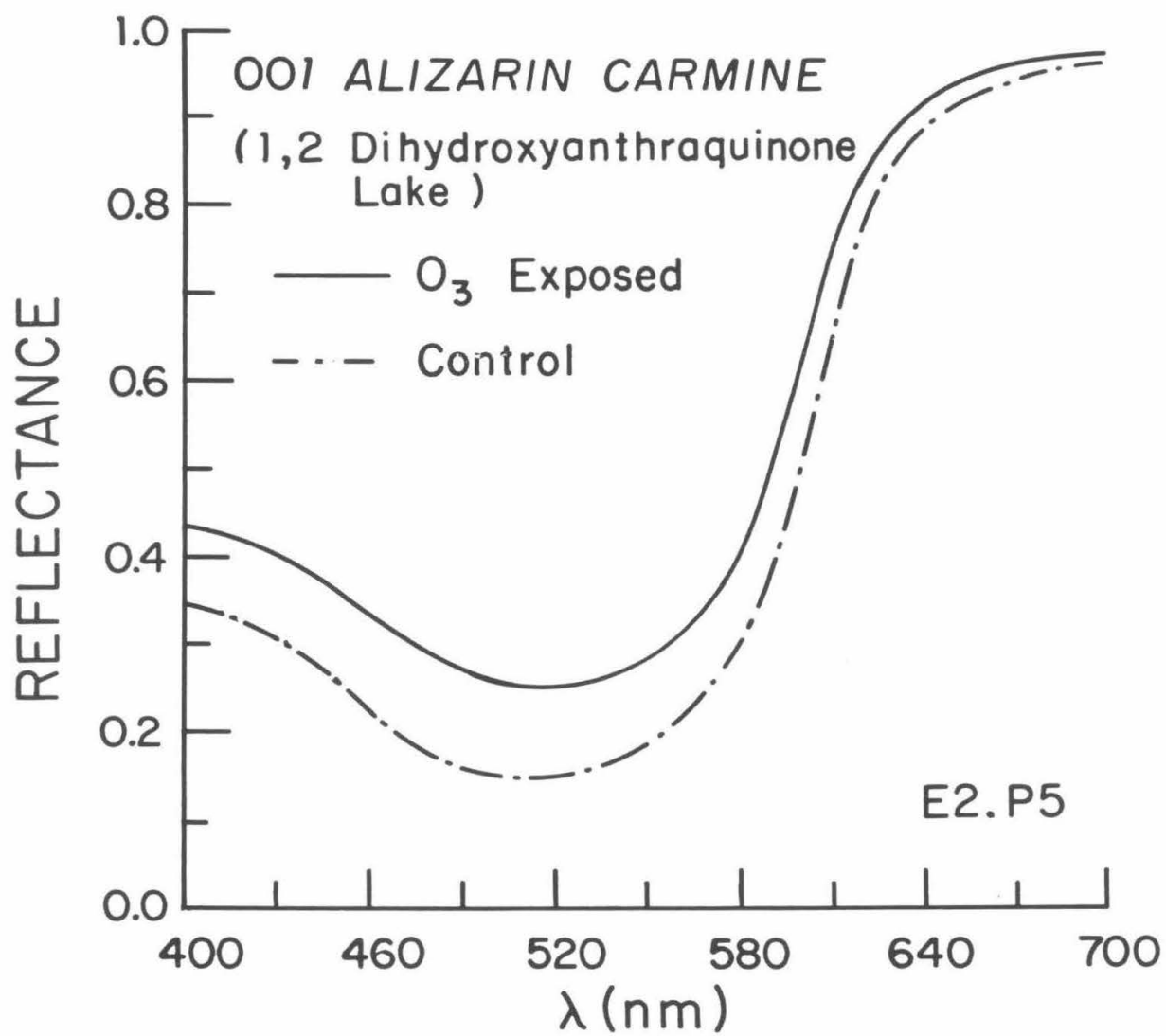


FIGURE 5



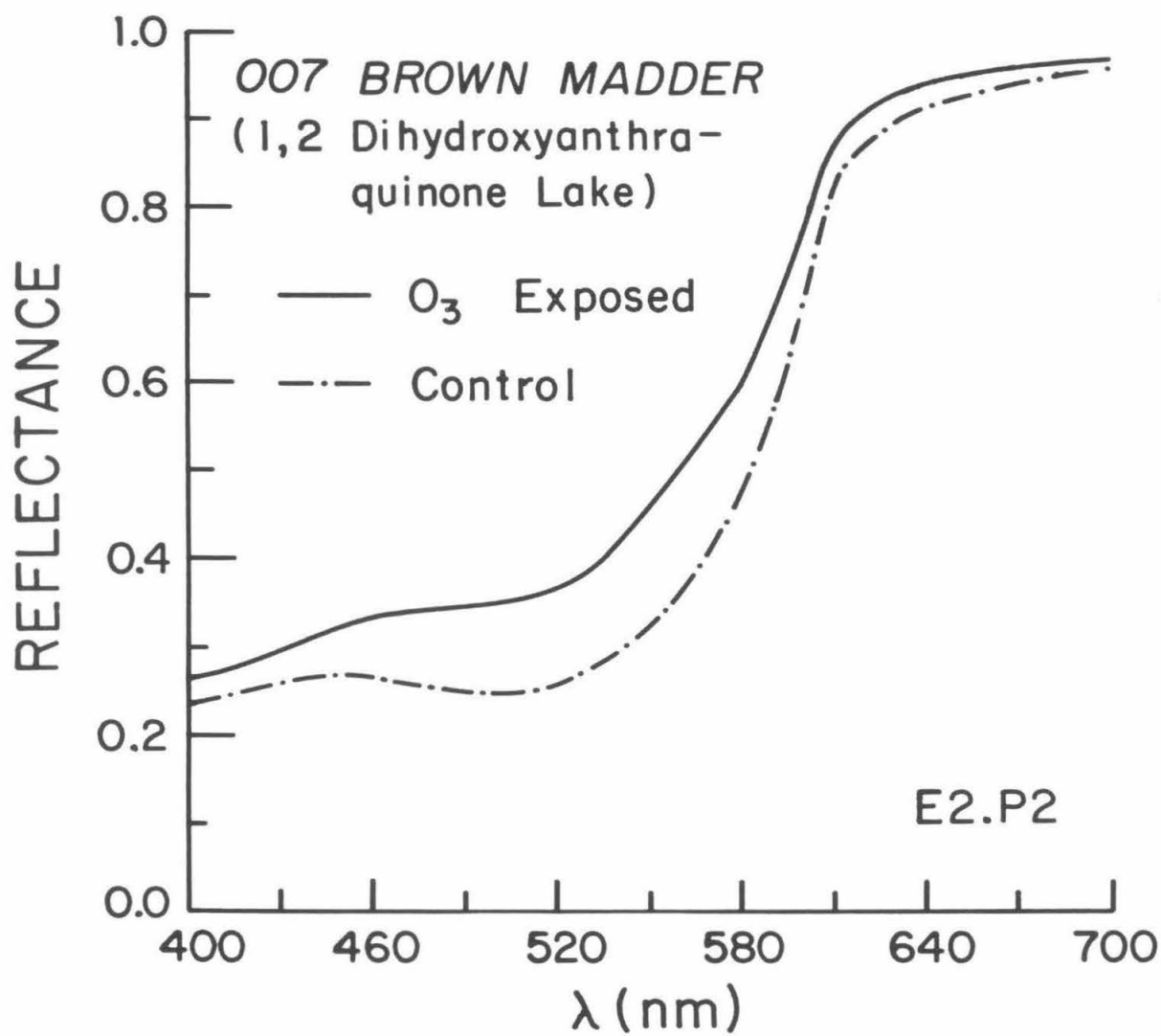
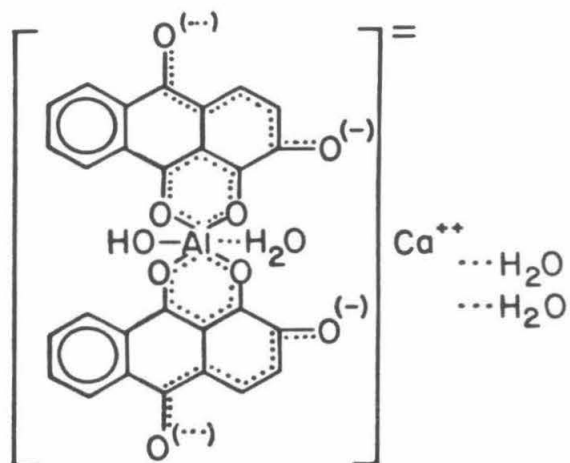
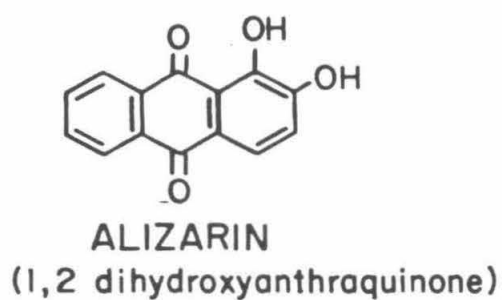
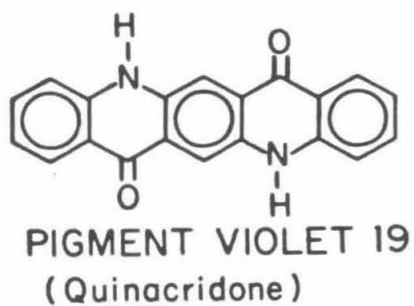
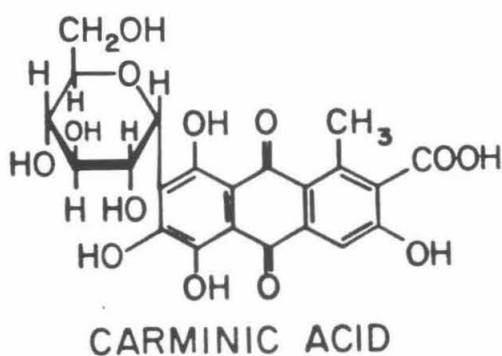


FIGURE 6



**AN ALIZARIN LAKE**  
(the calcium-aluminium lake  
shown as an example:  
 $\text{Ca} \cdot \text{Al}(\text{OH})(\text{Az})_2 \cdot 3\text{H}_2\text{O}$ )



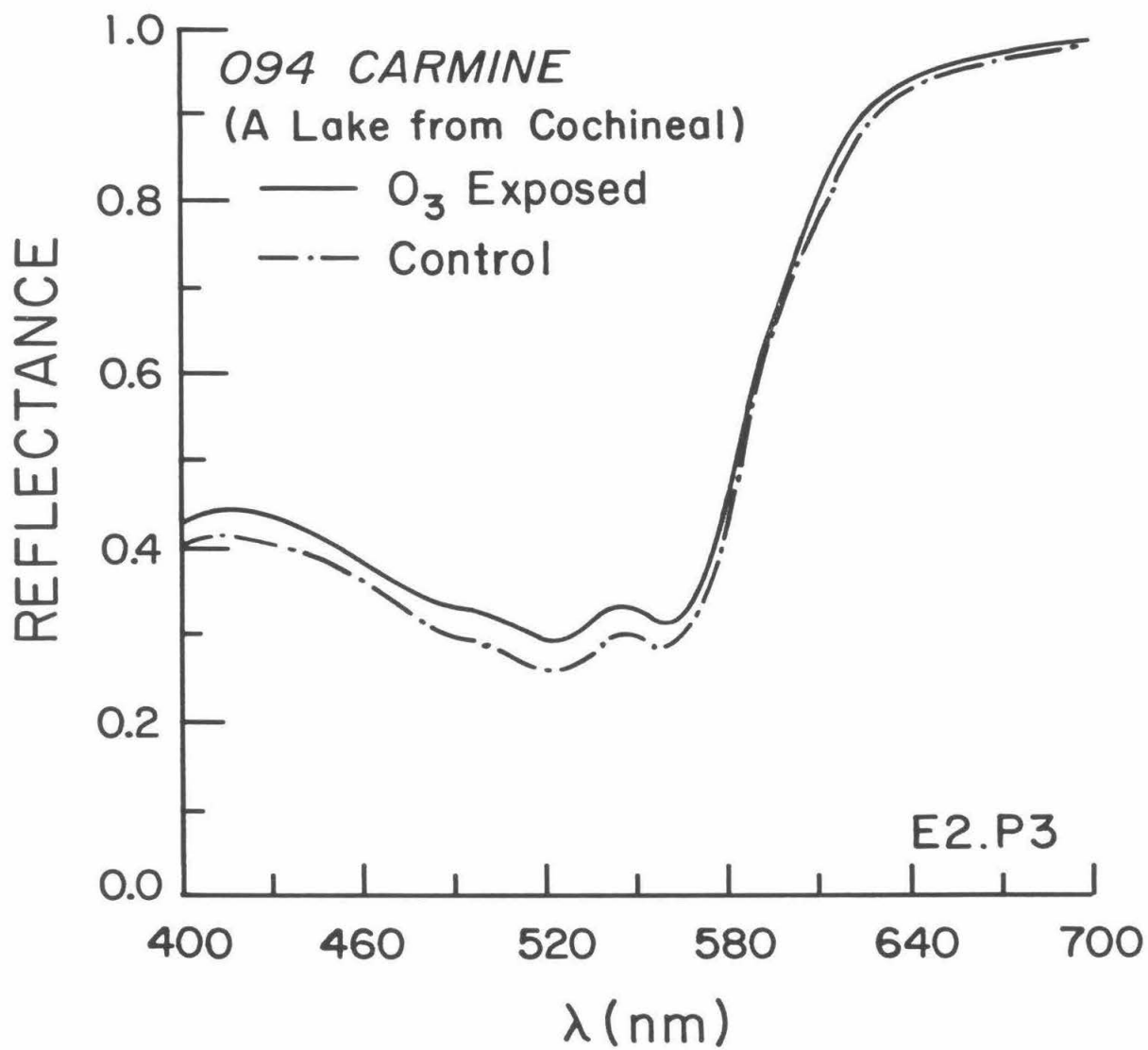


FIGURE 8

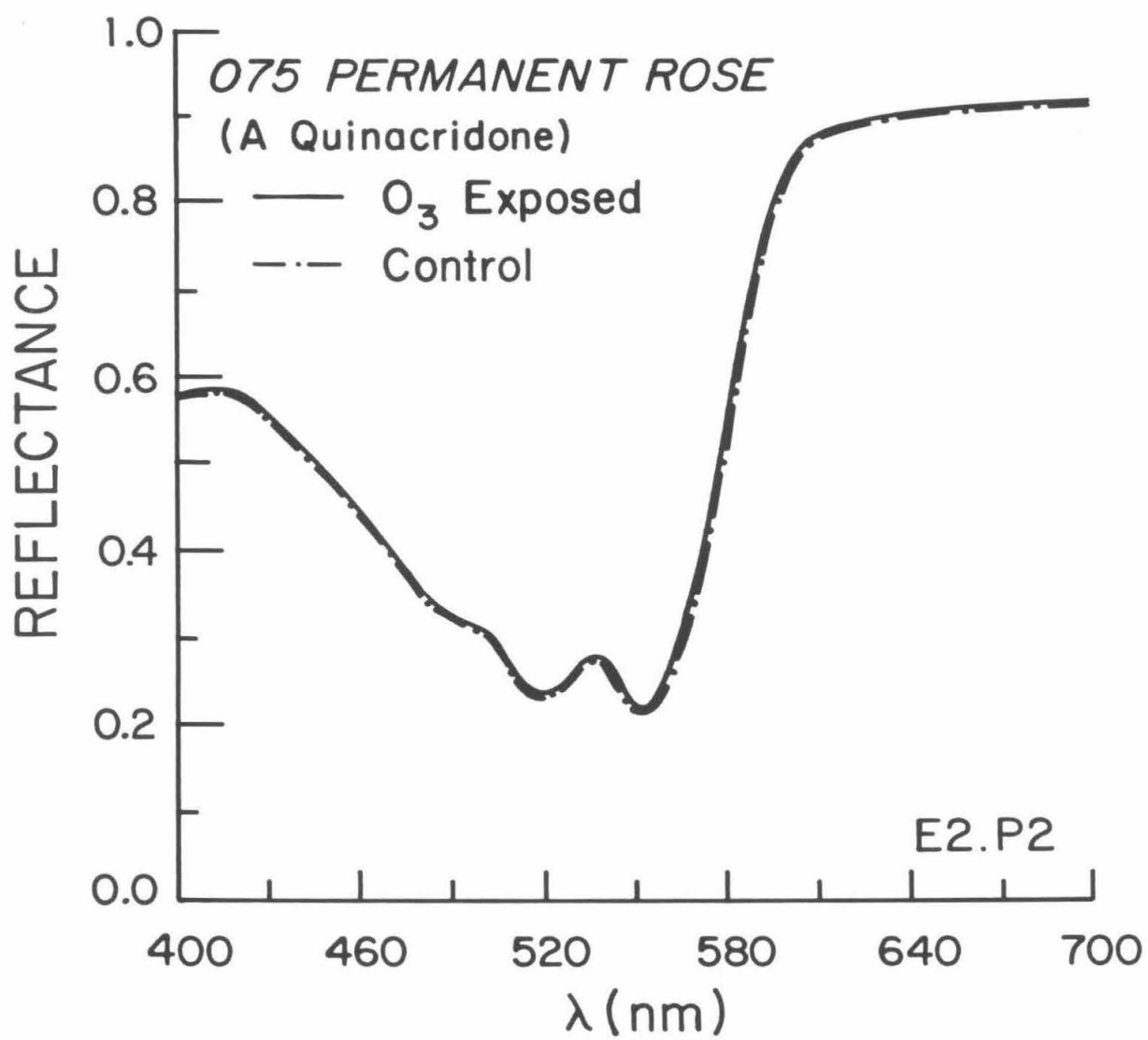


FIGURE 9

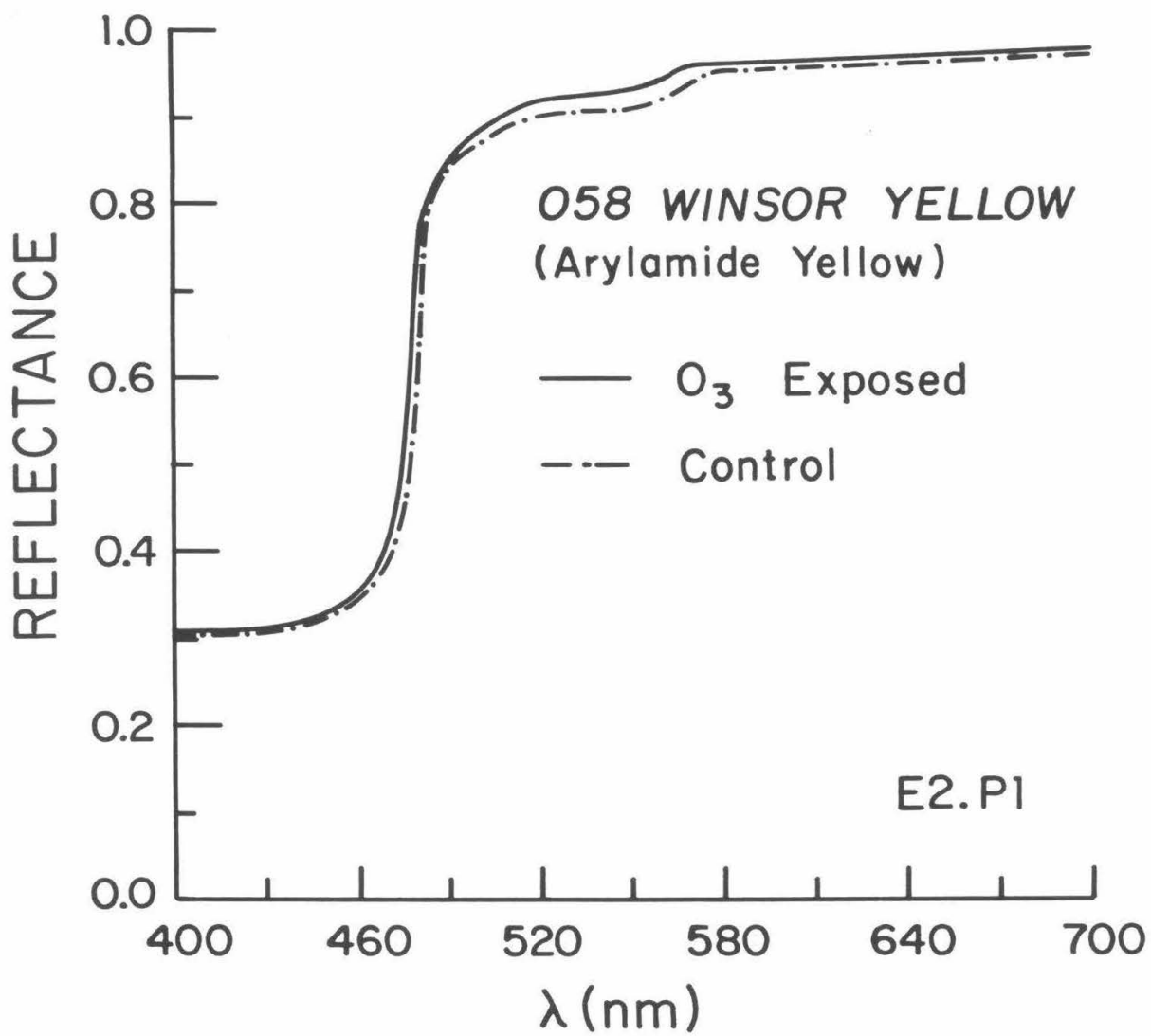


FIGURE 10

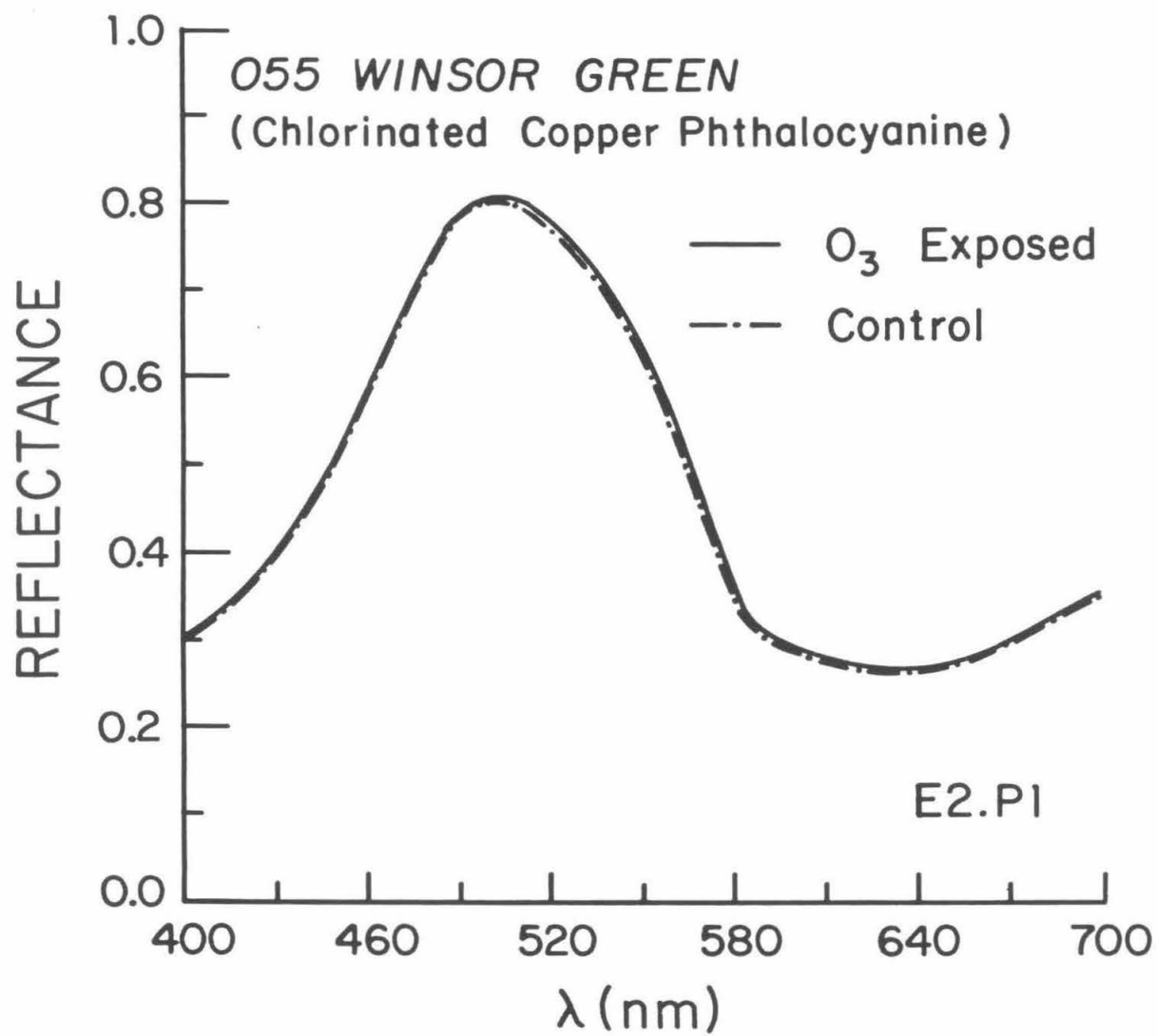


FIGURE 11

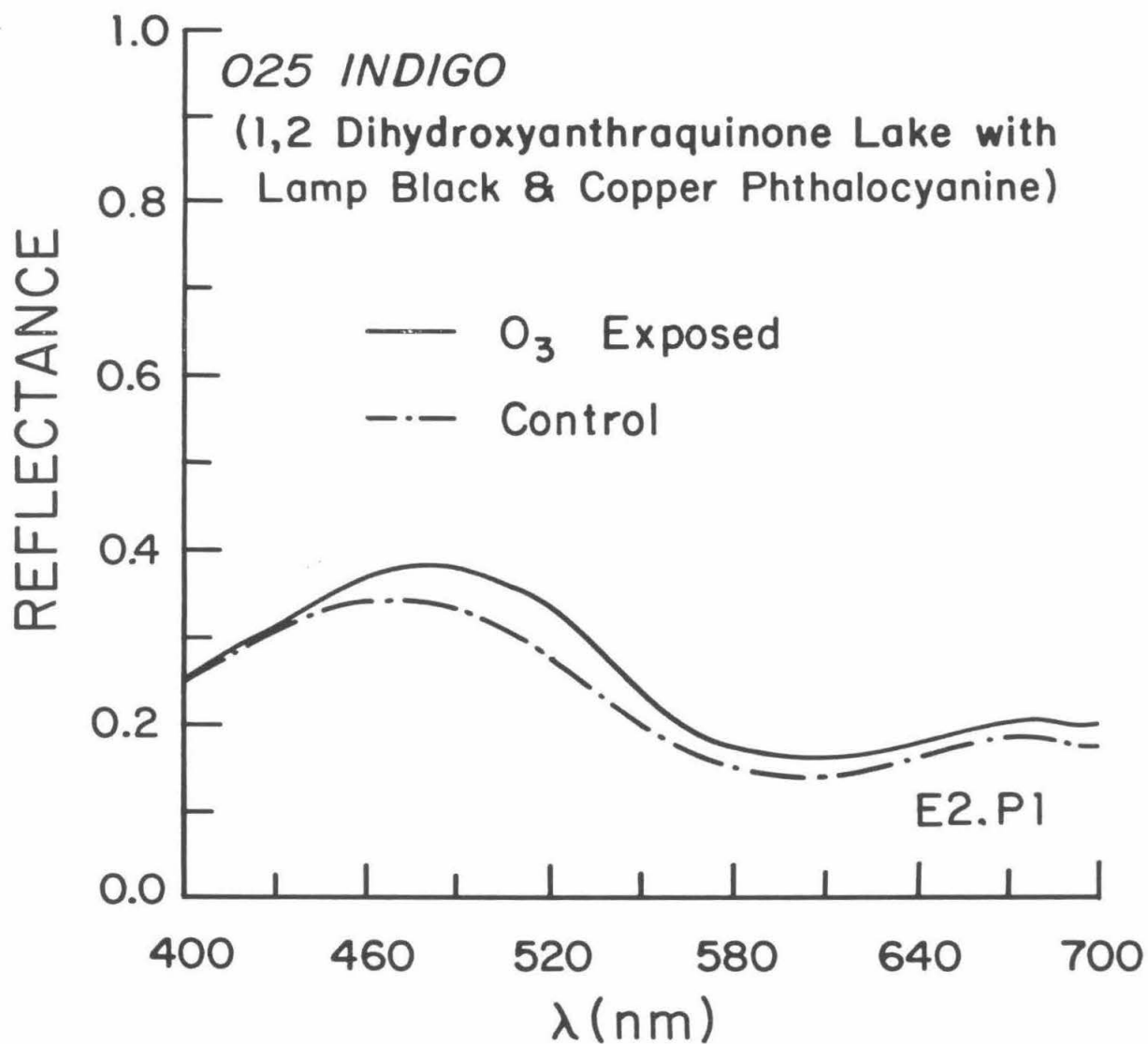


FIGURE 12

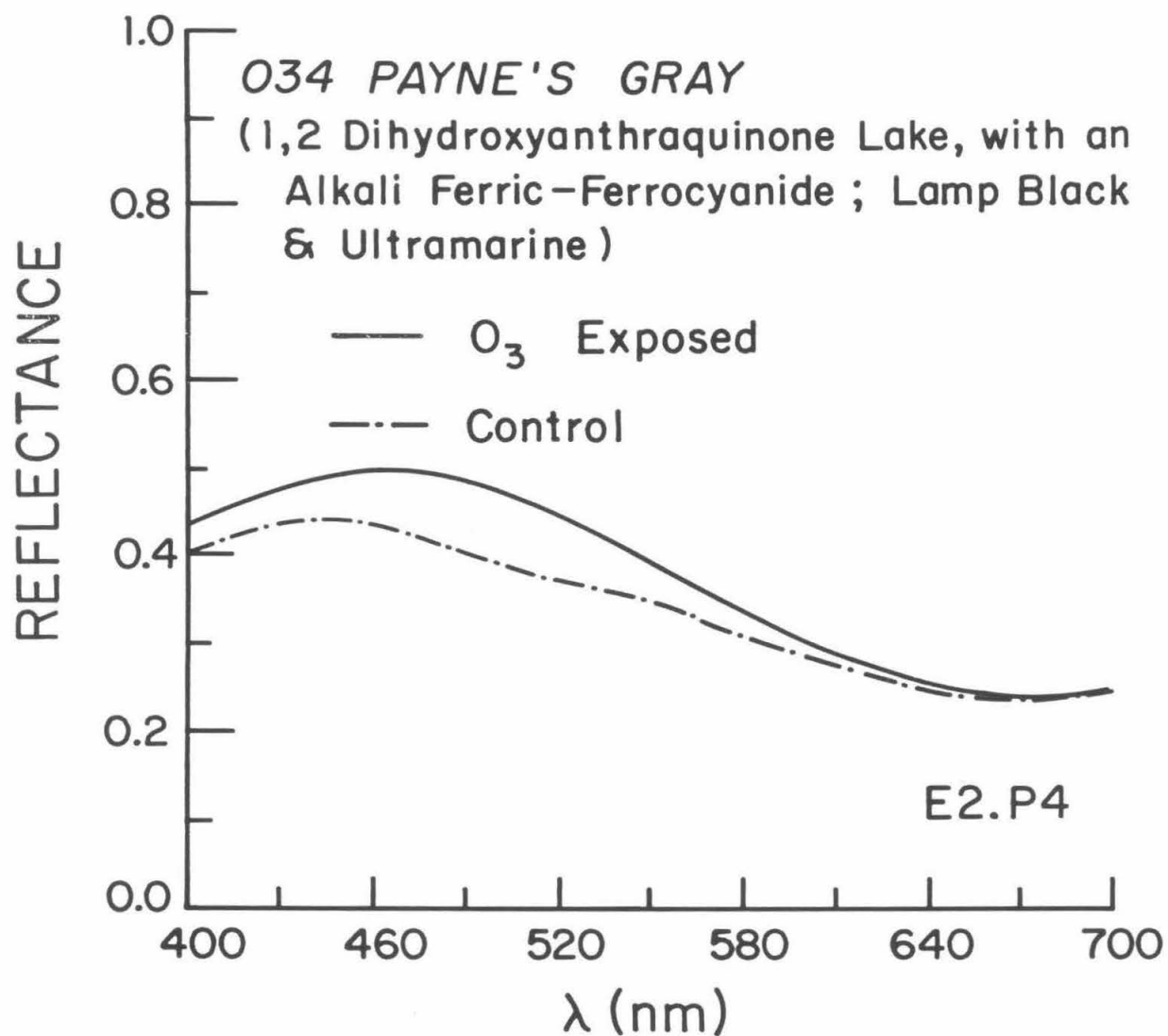


FIGURE 13



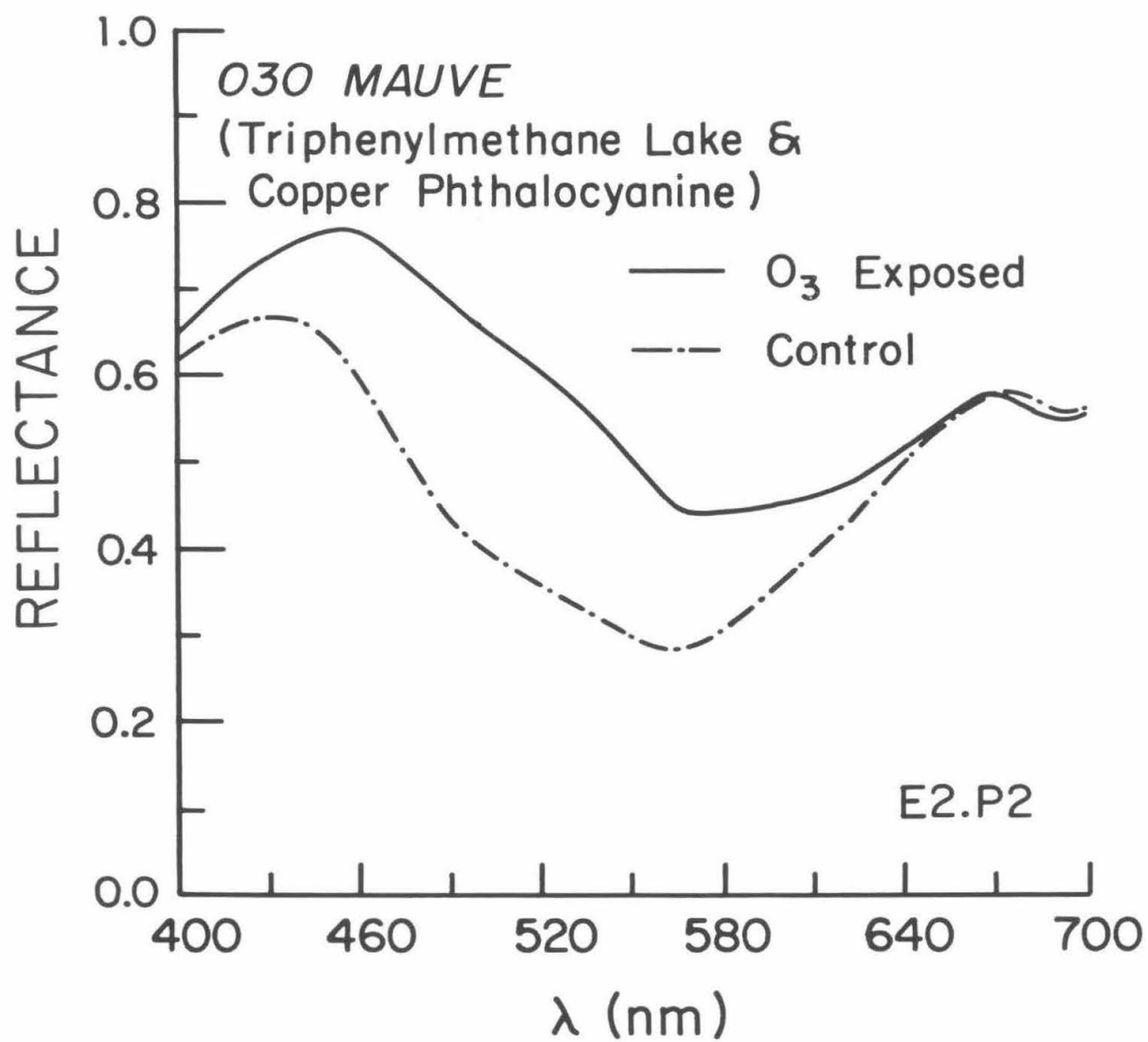


FIGURE 14

FIGURE 15 is a color slide (enclosed)